



INVESTOR IN PEOPLE

**CERTIFIED COPY OF
PRIORITY DOCUMENT**

101.003

The Patent Office
Concept House
Cardiff Road
Newport
South Wales
NP10 8QQ

09 1769 577

I, the undersigned, being an officer duly authorised in accordance with Section 74(1) and (4) of the Deregulation & Contracting Out Act 1994, to sign and issue certificates on behalf of the Comptroller-General, hereby certify that annexed hereto is a true copy of the documents as originally filed in connection with the patent application identified therein.

In accordance with the Patents (Companies Re-registration) Rules 1982, if a company named in this certificate and any accompanying documents has re-registered under the Companies Act 1980 with the same name as that with which it was registered immediately before re-registration save for the substitution as, or inclusion as, the last part of the name of the words "public limited company" or their equivalents in Welsh, references to the name of the company in this certificate and any accompanying documents shall be treated as references to the name with which it is so re-registered.

In accordance with the rules, the words "public limited company" may be replaced by p.l.c., P.L.C. or PLC.

Re-registration under the Companies Act does not constitute a new legal entity but merely subjects the company to certain additional company law rules.

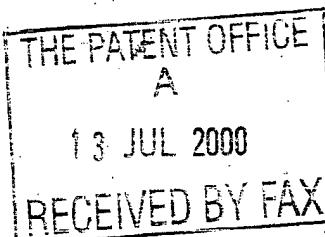
BEST AVAILABLE COPY

Signed *Andrea Jersey*

Dated 26 May 2004

1/77

Patents Form 1/77

Patents Act 1977
(Rule 16)

**The
Patent
Office**

13 JUL 2000 E352414-1 000254
P01/7700 0.00-0017127.6The Patent Office
Cardiff Road
Newport
Gwent NP9 1RH**Request for grant of patent**(See the notes on the back of this form. You can also get
an explanatory leaflet from the Patent Office to help
you fill in this form)

1. Your reference

32/46/46271 GB2

0017187.6

2. Patent application number
(The Patent Office will fill in this part)

13 JUL 2000

3. Full name, address and postcode of the or of
each applicant (*underline all surnames*)MEGGITT (UK) LIMITED
COWGROVE-WIMBORNE
DORSET
BH21 4EL

Patents ADP number (if you know it)

4274866003

If the applicant is a corporate body, give the
country/state of its incorporation

UNITED KINGDOM

4. Title of the invention

CATALYTIC REACTOR

5. Name of your agent (if you have one)

FITZPATRICKS

"Address for service" in the United Kingdom
to which all correspondence should be sent
(including the postcode)HOLBORN GATE, 1ST FLOOR
330 HIGH HOLBORN
LONDON WC1V 7QT

Patents ADP number (if you know it)

00000695001

6. If you are declaring priority from one or more
earlier patent applications, give the country
and the date of filing of the or of each of these
earlier applications and (if you know it) the or
each application number

Country	Priority application number (if you know it)	Date of filing (day/month/year)
GB	0001699.8	25/01/2000

7. If this application is divided or otherwise
derived from an earlier UK application, give
the number and the filing date of the
earlier application

Number of earlier application	Date of filing (day/month/year)
-------------------------------	------------------------------------

8. Is a statement of inventorship and of right
to grant of patent required in support of
this request? (Answer 'Yes' if:

- a) any applicant named in part 3 is not an inventor, or YES
- b) there is an inventor who is not named as an applicant, or
- c) any named applicant is a corporate body).

BEST AVAILABLE COPY

Patents Form 1/77

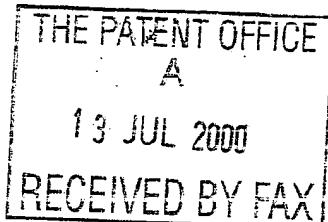
13. JUL. 2000 16:17

NO. 6678 P. 3

Patents Form 1/77

9. Enter the number of sheets for any of the following items you are filing with this form.
Do not count copies of same document

Continuation sheets of this form



Description	16
Claim(s)	
Abstract	
Drawing(s)	4 ✓ NO

10. If you are also filing any of the following, state how many against each item.

Priority documents

Translations of priority documents

Statement of inventorship and right to grant of a patent (Patents Form 7/77)

Request for preliminary examination and search (Patents Form 9/77)

Request for substantive examination (Patents Form 10/77)

Any other documents
(please specify)**BEST AVAILABLE COPY**

11.

I/We request the grant of patent on the basis of this application.

Signature

Date

13/07/2000

Fitzpatrick
(AGENT FOR APPLICANT)

12. Name and daytime telephone number of person to contact in the United Kingdom

ERIC EDE

TEL NO: 0141 306 9000

Warning

After an application for a patent has been filed, the Comptroller of the Patent Office will consider whether publication or communication of the invention should be prohibited or restricted under Section 22 of the Patents Act 1977. You will be informed if it is necessary to prohibit or restrict your invention in this way. Furthermore, if you live in the United Kingdom, Section 23 of the Patents Act 1977 stops you from applying for a patent abroad without first getting written permission from the Patent Office unless an application has been filed at least 6 weeks beforehand in the United Kingdom for a patent for the same invention and either no direction prohibiting publication or communication has been given, or any such direction has been revoked.

Notes

- a) If you need help to fill in this form or you have any questions, please contact the Patent Office on 0645 500505.
- b) Write your answers in capital letters using black ink or you may type them.
- c) If there is not enough space for all the relevant details on any part of this form, please continue on a separate sheet of paper and write "see continuation sheet" in the relevant part(s). Any continuation sheet should be attached to this form.
- d) If you have answered 'Yes' Patents Form 7/77 will need to be filed.
- e) Once you have filled in the form you must remember to sign and date it.
- f) For details of the fee and ways to pay please contact the Patent Office.

32 46271 GB2

1

CATALYTIC REACTOR

Field of the Invention

The present invention is applicable in the field of chemical engineering and especially relates to improvements in chemical reactors. In general the invention 5 relates to process control of temperature in a chemical reaction system and processing plant. Particularly the invention provides a reactor suitable for rapid conversion of a fluid reactant within the reactor in which the reactant temperature is maintained at a desired profile indirectly by means of a heat exchanging fluid.

Background of the Invention

In the majority of chemical processes there is a heat demand or a need to dissipate heat. Therefore, a wide range of chemical plant is involved in containing or conveying fluids which must at some stage of the process be either heated or cooled. One might consider furnaces, evaporators, distillation units, dryers and reaction vessels as plant where heat transfer manifests itself as a design and operational problem. In particular many industrial chemical processes employ reactors in which reactions are effected under given temperature and pressure conditions in the presence of a catalyst. Almost all these reactions generate or absorb heat i.e. they are exothermic or endothermic. The cooling effects for endothermic reactions generally adversely affect the rate of reaction and the corresponding parameters 10 such as conversion and selectivity of the products from the reaction. The uncontrolled heating of exothermic reactions generally leads to damage to the associated apparatus as the temperature can rise to a very high level. The reaction in such a case may become uncontrolled (so-called "run away reaction") and lead to unwanted by-products and undesired effects, such as deactivation of a process 15 catalyst. Furthermore, whilst an ideal catalyst does not theoretically participate in a reaction in reality many catalysts become degraded or poisoned as the reaction progresses and on an industrial scale the costs associated with catalyst regeneration or replacement represent a significant burden. It will be understood that such costs also must include the down time for the plant or restrictions on capacity if 20 a particular reactor has to be off-line for catalyst re-generation purposes. Therefore, it is desirable to prolong the life of a catalyst bed in view of the significant cost benefits 25 that may be obtained overall. The invention to be described hereinafter is ideally 30

32 46271 GB2

2

s suited to use in catalytic reactor design but can be adapted for other purposes. No distinction is made as to the application thereof to batch or continuous reaction systems.

Those skilled in the art recognise that it is beneficial for the changes in temperature resulting from the heating or cooling effects of the reactions to be controlled. It is well known that maintaining the temperature of the reaction at a given constant level may result in significant advantages to the reaction, such as improved conversion and selectivity, prolonged life of the catalyst and associated apparatus, reduced levels of unwanted by-products etc. In some cases, a slight varying of the theoretical temperature profile may be more beneficial.

In order to effectively control the temperature of reactions within an acceptable range, the chemical industry has devised several arrangements, those commonly used being discussed in standard references and texts e.g. one might consider the general teachings by Octave LEVENSPIEL in Chapter 19 of Chemical Reaction Engineering. The relative merits of each approach is also discussed therein.

Conventionally, the temperature inside reactors has been controlled by passing an auxiliary heat exchanging fluid through tubes or between plates, same forming a heat transfer conductive medium or thermal bridge whilst separating the reaction species from the auxiliary heat exchange fluid. Thus it will be understood that in such an indirect heat transfer system there is on the one hand a process path or zone and on the other an auxiliary fluid path or zone separated by the tube wall(s) or plate surface(s).

Considering this well known concept in relation to packed catalytic bed reactors, reactant fluid is passed through the catalyst bed and heat of reaction therein is controlled by contacting the catalytic bed reaction zone with such auxiliary fluid containing tubes or plates. However, particularly for highly exothermic reactions, such an approach has not been found to be ideal since the packed bed often develops heat gradients, e.g. the catalyst bed will be cooler at its areas of contact with the said tubes or plates and hotter within its depth remote from said tubes or plates, permitting formation of hot spots or moving hot fronts leading to variations in reaction progress within the bed as a whole. Thus at such a hot spot the reaction

32 46271 GB2

3

may proceed faster and hence catalyst therein will be more rapidly degraded. This will be particularly significant in the case of large plants.

Thus, it may be considered that the problems to be solved include the need to keep reactants and catalyst within a satisfactory temperature range with a view to 5 maximising reaction rate; minimising reactor or catalyst volume; maximising yield of desired products, minimising damage to catalyst (e.g. due to excessive heat, or direct contamination with liquid phases); and minimise by-product formation.

Typical approaches to such problems include the addition of quench gas to cool the system, but this leads to a loss in efficiency and may thus have an adverse 10 affect on yield. A further approach is to introduce a heat exchange step between adiabatic beds, which may involve the incorporation of heat exchangers into the reactor, but this leads to both design and operational problems due to the bulk of tubular designs and a lack of differential containment with plate exchangers. There are also problems with reactant re-distribution. Alternatively, the reactants may be 15 removed from the reactor for intermediate heat exchange e.g. cooling, but this also has an impact on plant design and process operation due to the additional expensive piping, and distribution problems with each extraction and re-injection. Thus this is not practical to do more than once or twice in any particular system.

A further approach is to adopt continuous heat exchange by packing tubes or 20 plates into the catalyst bed, but this leads to design inflexibility, additional expense, uneven packing of catalyst, and of course difficulty in replacing or regenerating the catalyst. Alternatively, heat exchange tubes, plates and passages of the reactor may be coated with catalyst, but here again this leads to an inflexible design, difficulties in applying catalyst reliably and restrictions upon available superficial catalyst surface 25 area. Also with this approach, there are obvious difficulties in replacing or re-regenerating the catalyst.

Fluidised bed reactors represent another potential solution to these problems, but these may not be ideally suited to all or certain reaction systems.

The possibility of using an inert or reactive diluent to ballast the temperature 30 of reactants in adiabatic beds has been considered but such a diluent must be heated, cooled, and pumped which places extra energy demands on the process and

32 46271 GB2

4

moreover, may also interfere with the intended reaction by presenting a diffusion barrier to reactants.

The problems may be further explained by considering the principles of the staged adiabatic packed bed reactor system which is an example of an arrangement designed to offer more control over the reactant temperature. This system uses an arrangement wherein a number of discrete, spaced apart zones of reaction are provided with means therebetween to control the temperature of the products leaving a first zone of reaction prior to entering the next reaction zone. No heat exchanging means is provided to control the temperature of the reaction in the zones of the reaction. Thus the reactant fluid entering the reactor at a desired temperature passes through a packed bed containing catalyst. Upon exiting this first stage, the reactant gas and any products will have a temperature higher or lower than that of the initial temperature depending upon the reaction thermal characteristics. A heat exchanger then heats or cools the reactant gas to a second desired temperature, which may or may not be equivalent to the temperature of the first, before passing to the next packed bed i.e. the second stage. This sequence is repeated until the desired conversion is obtained. Thus the temperature profile of the reaction will be stepped within an acceptable range of temperature, and will therefore not be truly isothermal.

An alternative proposal for a process and apparatus for controlling reaction temperatures is disclosed in US patent No. 5,600,053. This arrangement uses corrugated heat exchange plates spaced apart with each plate defining a boundary of a heat exchange flow channel on one side of the plate and a boundary of a reaction flow channel on the other. In the arrangement, a heat exchange fluid passes in the first of the aforementioned channels and a reactant stream passes through the second, preferably with a catalyst being present. This arrangement is intended to eliminate or minimise the typical step-wise approach to the so-called isothermal condition objective.

However, the arrangement proposed in US-A-5,600,053 requires adjacent corrugated plates to be joined together. For this purpose, smooth edges are provided to facilitate the assembly of superposed multiple plates to form channels. The plates are joined, such as by welding, along these smooth edges and hence the integrity of the seal of the channels formed by the corrugations in adjacent plates is

32 46271 GB2

5

not ideal, particularly where a large pressure differential exists between the heat exchange flow channels and the reaction flow channels since this will tend to urge the adjacent plates apart. This arrangement will thus place unnecessary constraints on parameters of the reaction, namely the relationship between the pressure of the 5 heat exchanging fluid and that of the reactant gas.

An earlier system is described in US-A-5,073,352 which proposes an apparatus for conducting a process of reforming gasolines, under low pressure and in the presence of at least one catalyst, in which heat required for the reaction is provided by a heat carrying fluid such as natural gas.

10 The apparatus described therein comprises a number of discrete reaction cells being arranged vertically and being of substantially parallelepipedic configuration. The cells are laterally spaced apart, thus forming channels therebetween for flow of the heat carrying fluid. The reforming catalyst-containing chambers are respectively either isothermal or adiabatic and dimensioned such that 15 height (H), width (W) and thickness (T) satisfy the conditions $H > W > T$, and H is at least twice the value of W, W lying in the range of 50 mm to 10,000 mm (0.05 – 10 metres) and T lies in the range of 2 mm 2,000 mm (0.002 – 2 metres). Thus there remains the possibility of hot spots and less than satisfactory thermal control in such large catalytic reactor volumes.

20 It is known to the man skilled in the art that the heat transfer coefficient in a packed bed is mainly dependent upon the catalyst particle size and the reactant fluid velocity through the catalytic bed. Unfortunately, both these parameters are process requirements and hence cannot be changed in order to improve the heat transfer coefficient in the packed bed, and hence in the reaction cells described in US 25 5,073,352. Additionally, it is difficult to move catalyst between narrow gaps or tubes, imposing limits on the dimensions of gaps or tubes through which catalyst is designed to flow.

Thus reactors of the known types according to the existing art have many significant limitations imposed on the heat transfer capability. Plate reactors offer 30 some advantages over tubular reactors on the auxiliary medium side, but the end result is not significant since the overall heat transfer coefficient is generally governed by the process side as discussed above. Tubular reactors on the other

13. JUL. 2000 16:19

32 46271 GB2

6

hand offer advantages over plate reactors with regard to mechanical capability, due to increased resistance to differential pressure between the reactant fluid and the heat exchanging fluid.

Considering the foregoing matters, it is an object of this invention to provide 5 improvements in chemical plant design and methods of operation thereof with a view to obviating or mitigating the drawbacks of the existing or previously proposed designs and methods.

Particularly, it is an aim of the present invention to provide an apparatus and 10 a process for the control of reaction temperature within an acceptable range during operation of the chemical process by an indirect heat transfer method using a heat exchanging fluid.

Another object of the invention to be described more particularly hereinbelow 15 is to provide an apparatus permitting control of the reactant temperature closely within a desired profile, more specifically, aiming to maintain the temperature at a substantially constant level i.e. to offer attainment of an isothermal reaction zone in so far as is practical on an industrial scale.

It is a further object of the invention to provide chemical plant which is improved over known plant equipment in terms of both cost and space efficiency considerations.

20

Summary of the Invention

The invention addresses the problems observed in the prior art by adopting the approach of staged adiabatic reactors and improving the performance thereof by design of a catalytic reactor comprising a catalyst bed and heat exchange means of the plate type in operative contact with the catalyst bed so as to receive reactants for 25 heat exchange purposes, wherein the heat exchange means is formed from a plurality of superposed metal plates wherein fluid flow channels have been chemically milled e.g. by etching, according to a pre-determined pattern, said channel-bearing plates being aligned during superposition to define discrete heat exchange pathways for fluids and diffusion bonded together. The plate exchangers 30 to be adopted are panels made according to a chemical milling or etching technique

13. JUL. 2000 16:19

32 46271 GB2

7

commonly referred to as printed circuit design since the manner of fluid channel definition and formation is analogous to the manufacture of printed circuit boards.

Ideally, multiple heat exchange panels are embedded within the catalyst bed the design being such that the contact face area of the panels is similar to the contact 5 face area of the catalyst bed. A plurality of such catalyst beds can be arranged in succession having a heat exchange panel arranged between each bed, typically providing at least 3 such beds in series.

The heat exchanger of choice is one formed from a plurality of plates superposed and diffusion bonded to form a stack of plates, wherein fluid channels 10 are defined in said stack by virtue of a pre-treatment of said plates wherein each plate is selectively configured according to the desired pattern of channels by a chemical treatment to remove surface material e.g. by chemical etching, to a desired depth. Such a pre-treatment of the plates is conducted in a manner analogous to manufacture of printed circuit boards (PCBs) and for this reason the reactor design 15 described herein can be described as a printed circuit reactor (PCR). Likewise, the heat exchanger thus formed for inclusion in the reactor may be referred to as a printed circuit heat exchanger (PCHE).

The proposed reactor design offers an infinite variety of auxiliary heat exchanging fluid and reactant fluid pathways of very small dimensions which enables 20 significantly enhanced process control. Typically the passages are of very small bore, typically of less than about 3 mm in depth. The nature of the design is such that it lends itself to construction of heat exchangers in small sub-panel units that can be readily fastened together, e.g. by welding. Furthermore, it is possible to provide designs of reactor in which the PCHE panel thickness differs at different stages of the 25 reactor, and to vary the catalyst bed thickness stage to stage. The composition of the catalyst in each stage may be the same or varied according to process requirements with a view to enhancing production.

Preferably, all the heat exchangers used in the reactor are panels that are entirely of the printed circuit heat exchanger type (PCHE). Typically in such an 30 arrangement, the heat transfer dimensions are smaller than catalyst particle dimensions, ensuring that the temperature profiles inherent in heat transfer to fluids in passages are not significant relative to the catalyst particle size. Also, the

dimensions of the heat transfer are relatively small to the bed depth, so that any passage-scale temperature profiles occupy only a very small proportion of the individual catalyst bed lengths, e.g. typically up to about 200 mm. This contrasts significantly with the prior art use of exchanger tubes of say 25 mm outer diameter, 5 which essentially cause downstream wakes in the temperature profiles which are then necessarily of a scale that is significantly larger than the individual catalyst particles and extend across at least a significant proportion of each catalyst bed.

The catalyst may be of variable form, e.g. selected from spherical, cylindrical, hollow bodies, solid particles, expanded or porous solids, coated matrix catalyst or 10 the like supported catalysts. Commonly particles of up to about 10 mm (major dimension) are contemplated.

Preferably, the potential for catalyst particles to enter the passages of the PCHE is restricted by provision of a screen ideally of a durable mesh capable of restraining particles of catalyst at operational temperatures.

15 The heat transfer medium may be a gas, or a liquid, either boiling or condensing according to the process (exothermic or endothermic), and it is considered that reactants may be used as a heat transfer medium during the reaction process, either prior to entry to the catalytic reaction zone enabling a reactant pre-heat step or after exiting the reactor as a reactant/product cooling step.

20 The PCHE panel may be of a thickness intended to minimise pressure drop between catalytic zones, e.g. up to about 100 mm. This also enables matching of heat transfer area of PCHE and catalyst bed volume to achieve a cost-effective design, which is difficult to achieve with the prior art coated passage designs or in packed plates and tube designs.

25 The PCHE panel design enables variable passage length and configuration, e.g. tortuous pathways with convolutions and or zigzags to enhance heat transfer, thereby permitting closer approach of reactant and heat transfer medium temperatures, and offering temperature profiles in the system which are more consistent with design parameters.

30 The invention further addresses the drawbacks of the known art by providing a process for conversion of a fluid reactant which process according to the invention

32 46271 GB2

9

uses a catalytic reactor comprising catalyst bed and heat exchange means of the plate type in operative contact with said bed and having discrete fluid pathways for heat exchange between fluids at differing temperatures whilst avoiding mixing of the fluids, the said process providing the appropriate fluid reactant species to be
5 converted in a catalytic reaction zone in the catalyst bed within the reactor and at a predetermined stage of reaction introducing at least a portion of the fluid reactant species into a reactant fluid pathway within said heat exchange means, and also introducing an auxiliary fluid at a temperature differing from that of the fluid reactant species into another fluid pathway within said heat exchange means and juxtaposed
10 to the first whereby the discrete nature of the respective pathways permits indirect heat exchange between the fluid reactant species, said process being optionally repeated in successive stages.

According to a modification of the process additional fluid reactant species may be introduced at subsequent catalyst bed stages. Thus although it is envisaged
15 that the process can be operated in a series of stages in the manner of the known staged adiabatic reactor systems the proposed reactor design permits greater control over the process not only in terms of heat management but also in terms of chemical reaction control.

Thus according to one aspect of the present invention, there is provided an
20 apparatus for controlling the temperature profile of a reactant fluid in the presence of a catalyst during an endothermic or exothermic chemical reaction, comprising a reactor having reactant fluid inlet means and reactant fluid outlet means; catalytic beds being provided therebetween, spaced apart by a printed circuit heat exchanger (PCHE); said heat exchanger comprising heat exchanging fluid inlet means, heat
25 exchanging fluid outlet means, a first channel or set of channels for passage of the heat exchanging fluid, and a second channel or set of channels in communication with the adjacent catalytic beds to allow passage of the reactant fluid from one catalytic bed to the next, said second channel or set of channels not being in communication with the reactant fluid.

30 Preferably a screen made of a fine mesh lines the walls of the catalytic bed, and said walls are ideally formed at least in part by plates of the said heat exchangers. The mesh acts to resist migration of catalyst into the reactant fluid-receiving channels of the heat exchanger down stream of the catalytic reaction zone.

32 46271 GB2

10

The or each heat exchanger stack may be formed from a length or block of superposed plates by division e.g. by cutting into individual slices of a desired dimension which enables very slim designs of great strength.

Thus in one construction, a first such channel or set of channels is perpendicular to a second such channel or set of channels. In an alternative construction, the respective channels are parallel. Naturally one would generally arrange the construction such that juxtaposed channels contain respectively reactant fluid species on the one hand and auxiliary fluid on the other to achieve the desired heat transfer. In this way temperature control is achieved indirectly without mixing of the reactant fluids with the auxiliary fluid media.

The PCHE panel design may include plates having passages etched on one side or both sides, and the panel may comprise an assembly of stacked plates consisting of un-etched plates (blanks) with suitably juxtaposed etched panels to form a desired passage assembly in the final panel. The stacked plates thereby form a laminar assembly of superposed metal plates wherein fluid flow channels are arranged according to a pre-determined pattern, said channel-bearing plates being aligned during superposition to define discrete heat exchange pathways for fluids, and the assembly is formed into a unitary heat exchanger panel by a diffusion bonding technique.

The profile of the channels, i.e. cross-sectional profile perpendicular to the flow path, is generally not critical but curved shapes are conventionally used and are relatively easy to form by chemical milling but other profiles could be adopted if desired by use of an appropriate tool in combination with the chemical process.

It will be understood that the plates having the appropriate channels defined by etching or the like are stacked and diffusion bonded together to form heat exchanger panels, and that such panels so formed can, if necessary, be juxtaposed and joined e.g. by welding to provide a larger panel of a desired height and width to match the required catalyst bed cross-sectional area. Use of blank (un-etched plates) is appropriate in some instances to complete a panel and close the open side of channels formed in an adjacent etched plate. The reference to panels is for convenience and is not intended to indicate a dimensional limitation. However, it will be appreciated that the dimensions of the heat exchanger unit will vary according to

3.2 66271 GB2

11

a chosen reactor design, and that currently available manufacturing equipment may impose some practical limitations on panel size in one step. If it is desired to form relatively large size panels, such practical limitations can be readily overcome by juxtaposition of a plurality of panels of a size formed within the capacity of the 5 available equipment and joining thereof by a suitable method such as welding. In this way a variety of shapes and sizes of the PCHE panel can be constructed.

In use of such a PCR reactor, the heat exchanging fluid may be caused to flow in a direction substantially perpendicular to the flow of the reactant stream. Alternatively, the directions of flow may be substantially parallel and either co or 10 counter current according to the operators choice taking account of the reaction process to be controlled.

The invention is primarily intended for use with reactions using heterogeneous catalyst systems.

Depending on the particular reaction, optimisation thereof may be obtained by 15 providing more than one catalyst, and in particular by providing different catalysts in separate catalytic beds.

The heat exchanging fluid may be a liquid or gas, as is understood by those in this art. Such fluids typically include molten salts, molten metals or hot water to provide liquid auxiliary media or may be hot gases, steam or superheated steam 20 whereby heat may be indirectly added to a reaction system. In the converse case chilled liquids or gases may be used. Chemical engineers will readily consider the wide range of auxiliary working fluids available and be aware of both sensible heat and latent heat considerations in matching the auxiliary fluid needed to the process demands of the reaction for conversion of the fluid reactant species.

25 Each heat exchanger stack or each auxiliary fluid media channel, or set of channels, may contain a different auxiliary fluid to optimise the temperature profile of the reactant fluid within the reactor.

According to another aspect of the invention, there is provided additional 30 means to make the aforementioned reactor specifically suitable for use as a moving bed reactor, namely catalyst inlet means, catalyst outlet means and means for feeding new or regenerated catalyst into the catalyst inlet means, and further means

32 46271 GB2

12

to remove catalyst from the catalyst outlet means. Preferably catalyst is allowed to progress towards the catalyst outlet under the influence of gravity. The man skilled in the art will be aware of alternative manners in which moving bed reactors may operate and the scope of the invention should not be restricted to the particular 5 method described herein.

In the case of such a moving bed reactor, the bed width is a multiple of the catalyst diameter preferably at least 3 times that diameter.

According to a still further aspect of the invention, there is provided a process for indirectly controlling the temperature profile of a reaction fluid in the presence of a 10 catalyst during an endothermic or exothermic chemical reaction, comprising passing a reactant fluid from a reactant fluid inlet means in a reactor to a first catalytic bed before passing through a first channel or set of channels in a printed circuit heat exchanger (PCHE) and subsequently passing to a second catalytic bed; passing a heat exchanging fluid from a heat exchanging inlet means to a heat exchanging 15 outlet means through a second channel or set of channels in the said printed circuit heat exchanger (PCHE); and exchanging heat between the heat exchanging fluid and the reactant fluid whilst passing through the said printed circuit heat exchanger (PCHE); the products of the reaction leaving the last catalytic bed being passed to a reaction fluid outlet means.

20 In an alternative embodiment, the process for controlling the temperature profile of a reaction fluid in the presence of a catalyst during an endothermic or exothermic reaction further comprises passing catalyst through the catalytic bed, catalyst leaving the bed being replaced by new or regenerated catalyst.

Preferably the catalyst is a heterogeneous catalyst.

25 In a preferred embodiment, the heat exchanging fluid flows in a direction substantially perpendicular to the flow of the reactant stream. Alternatively, the directions of flow may be substantially parallel and either co or counter current.

The heat exchanging fluid may be liquid or gas, as is recognised already in the prior art. Such fluids include molten salts and boiling water.

13. JUL. 2000 16:21

32 46271 GB2

13

It will be recognised that the invention in its broadest aspect provides a staged reaction system containing successive chemical reaction zones and heat transfer zones, the latter containing plate heat transfer surfaces bearing micro-channels etched therein according to a pre-determined pattern, and offering not only 5 the optimum indirect heat transfer strategy but also the ability to individually prepare fluid reactants thermally for the next reaction zone by selection of the inlet channel and its relationship to adjacent auxiliary fluid channels. The system can be specifically designed to handle substances of different volatilities.

Description of the Drawings

10 Embodiments of the invention will be described hereinbelow with reference to the accompanying drawings in which:

Figure 1 is a sectional side view of part of a reactor in accordance with the present invention;

15 Figure 2 shows the temperature profile for a highly exothermic reaction demonstrating "hot-spot" problems;

Figure 3 in contrast to Figure 2 shows the temperature profile for an exothermic reaction controlled by means of a tubular reactor designed in accordance with the present invention; and

20 Figure 4 shows a printed circuit reactor in side elevation (Fig 4(a)), and in end elevation (Fig. 4(b)).

Mode for Carrying out the Invention

Referring to the drawings, Figure 1 shows a reactor 1 provided with a reactant fluid inlet 2 and a reactant fluid outlet (not shown) through which a reactant fluid to be processed is passed.

25 The reactor 1 comprises at least one catalytic bed 4. The catalytic bed 4 is substantially vertical and the chamber walls defined therefor are preferably smooth. These smooth walls facilitate the movement of the catalyst through the chamber where the apparatus is configured as a moving bed reactor.

Provided adjacent to the catalytic bed reactor 4 are printed circuit heat 30 exchangers (PCHE) 5. The PCHEs 5 have at least two sets of channels formed

32 46271 GB2

14

therein, a first set of channels 6 provided for passage of the reactant fluid from a first catalytic bed 4 to a second, and a second set of channels 7 through which the heat exchanging fluid flows. The first of the PCHEs 5, in communication with the reactant fluid inlet 2, is larger than the other PCHEs and contains pre-heat channels 3. These 5 pre-heat channels 3 are extra vertical passages in which heat exchanging fluid flows and heats the reactants to a suitable temperature for reaction prior to the reactants entering the first catalytic bed 4. In this example the pre-heat channels 3 make three passes across the reactant flow, although this is an arbitrary figure.

An optional screen of a fine mesh suitably dimensioned according to the size 10 of the catalyst particles is provided in this embodiment to cover the ends of the reactant fluid channels adjacent to the catalytic beds to prevent migration of catalyst into said channels, especially the down stream channels with respect to the catalyst to reduce the risk of blockages inhibiting flow.

Flow performance can be optimised and blockages decreased inside the 15 channels, if the channel walls are smooth and free of crevices. The channels are formed by joining individual plates together, said plates having channels formed in at least one surface, such as by a milling process. In order to ensure the crevice free design, it is thus important that any joining process used to join adjacent plates together does not interfere with the machined channels. This therefore limits the 20 acceptability of use of conventional welding processes and exposes the weakness of prior art proposals such as US-A-5,600,053. However, diffusion bonding processes, wherein the plates are placed under pressure and heated close to the melting temperature of the plate metal thereby encouraging grain growth across the boundary, allows such a crevice-free design. This also enables the plates to be 25 joined adjacent each of the channels, increasing the mechanical capabilities of the channels and allowing greater pressure differentials between the pressure of the reactant fluid and the heat exchanging fluid. This design of heat exchanger has been proven by the designers of the proposed PCR since 1985 when Heatic first introduced its compact printed circuit heat exchangers (PCHEs). The application of 30 the diffusion bonding technique is now understood in the art.

The design of such heat exchangers also facilitates a greater rate of heat exchange in a given volume, reducing the space requirements for a given reaction.

32 46271 GB2

15

Thus incorporating them in a custom reactor design offers hitherto unforeseen advantages.

- Depending on the performance requirements of the reactor 1, additional catalytic beds 4 may be provided, spaced apart by additional heat exchangers 5.
- 5 The final catalytic bed 4 in the series is in communication with the reactant fluid outlet means to enable the exit of the products of the reaction from the reactor 1.

In the alternative embodiment wherein the apparatus is adapted to be suitable for use as a moving bed reactor, catalyst outlet means (not shown) are provided adjacent the lower end of each of the catalytic beds 4, through which the catalyst is urged by means of gravity. The catalyst leaving the catalytic beds 4 may be fed to a regenerator and then passed through catalyst inlet means (not shown) provided adjacent the top of each of the catalytic beds 4. Alternatively, new catalyst material may be passed through the catalyst inlet means, consequent to exit of catalyst through the outlet means.

- 15 Thus the apparatus of the invention facilitates a series of adiabatic reactions, the temperature of the reactant stream being altered between successive reactions to maintain the reaction temperature within an acceptable range and thus conform to the desired temperature profile.

Figure 2 illustrates a poorly controlled exothermic reaction (phthalic anhydride production) with tubular inlet temperature showing severe hot spot development. Excessive inlet temperature leads to thermal runaway which under these hot-spot limited conditions leads to restriction of o-xylene feed to significantly less than 100g/Nm³. In contrast, a reactor containing 24 catalyst beds including PCHE stages between catalyst beds enables a very controllable process having the temperature profile illustrated in Figure 3. This shows temperature build up in the catalyst bed control (process development left to right) as an inclination and heat exchange by a sharp declination. A significant safety margin over thermal runaway is maintained by low inlet temperature and short catalyst beds (about 125 mm), even with in excess of 100g//Nm³ o-xylene in the feed. However, the average temperature gradually rises in a controllable and predictable manner as the reactants progress through successive catalytic beds to enable increase reaction rate in later stages where the threat of thermal runaway is much reduced.

32 46271 GB2

16

The number of heat exchangers to be used is calculated in accordance with methods known to the man skilled in the art in order to avoid hotspots and to allow the process to run at a higher load of feed per unit of flow rate.

Figure 4 illustrates a printed circuit reactor 11, suitable for use in the production of phthalic anhydride, and having a reactant fluid inlet 12 for receiving reactant fluid to be processed and a fluid outlet 13 through which a product can be recovered.

The reactor 11 comprises at least one catalytic bed 14 in communication with the reactant fluid inlet 12. The catalytic bed 14 is substantially vertical and the chamber walls defined therefor are preferably smooth. These smooth walls facilitate the movement of the catalyst through the chamber where the apparatus is configured as a moving bed reactor. Provided adjacent to the catalytic bed reactor 14 are printed circuit heat exchangers (PCHE) 15.

13-07-00 16:23

01413533932

P. 20

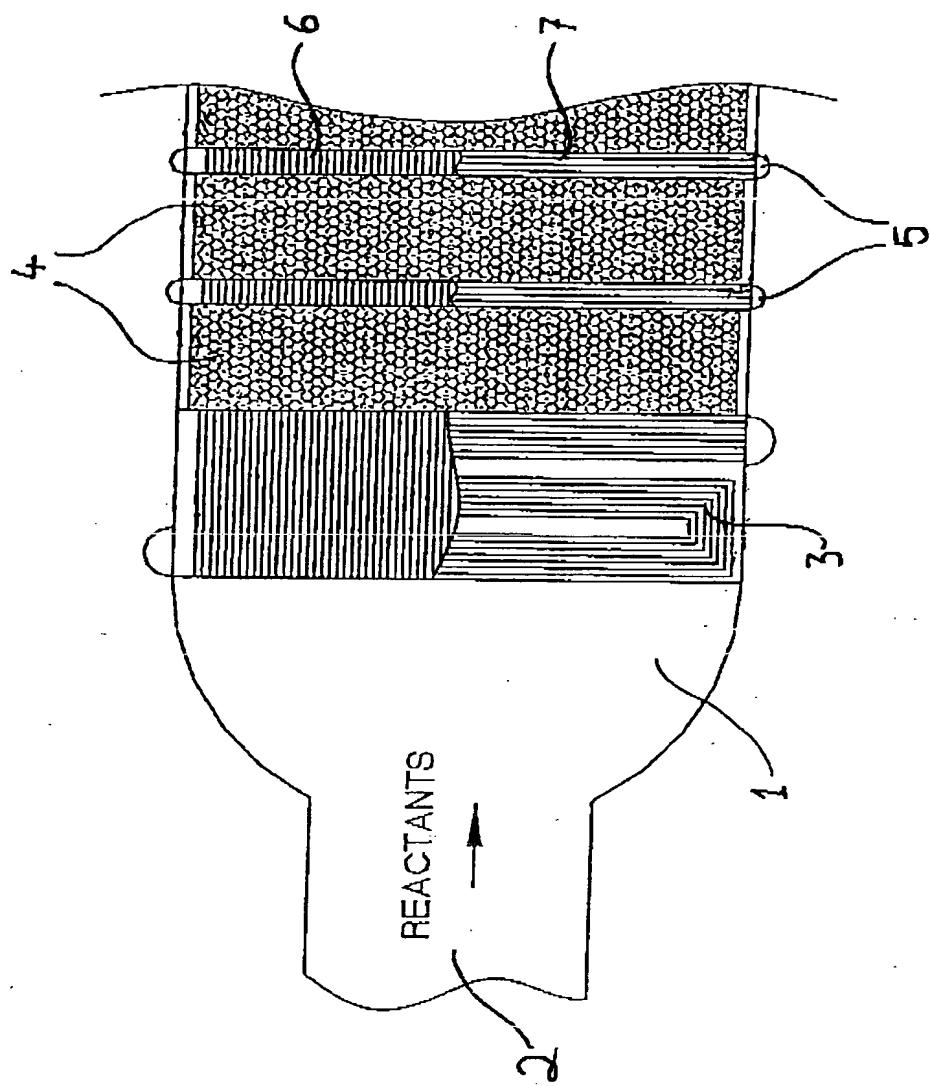
R-925 Job-265

13. JUL. 2000 16:22

NO. 6678 P. 20

1/4

Fig 1.



13-07-00 16:23

01413533932

P.21

R-925 Job-265

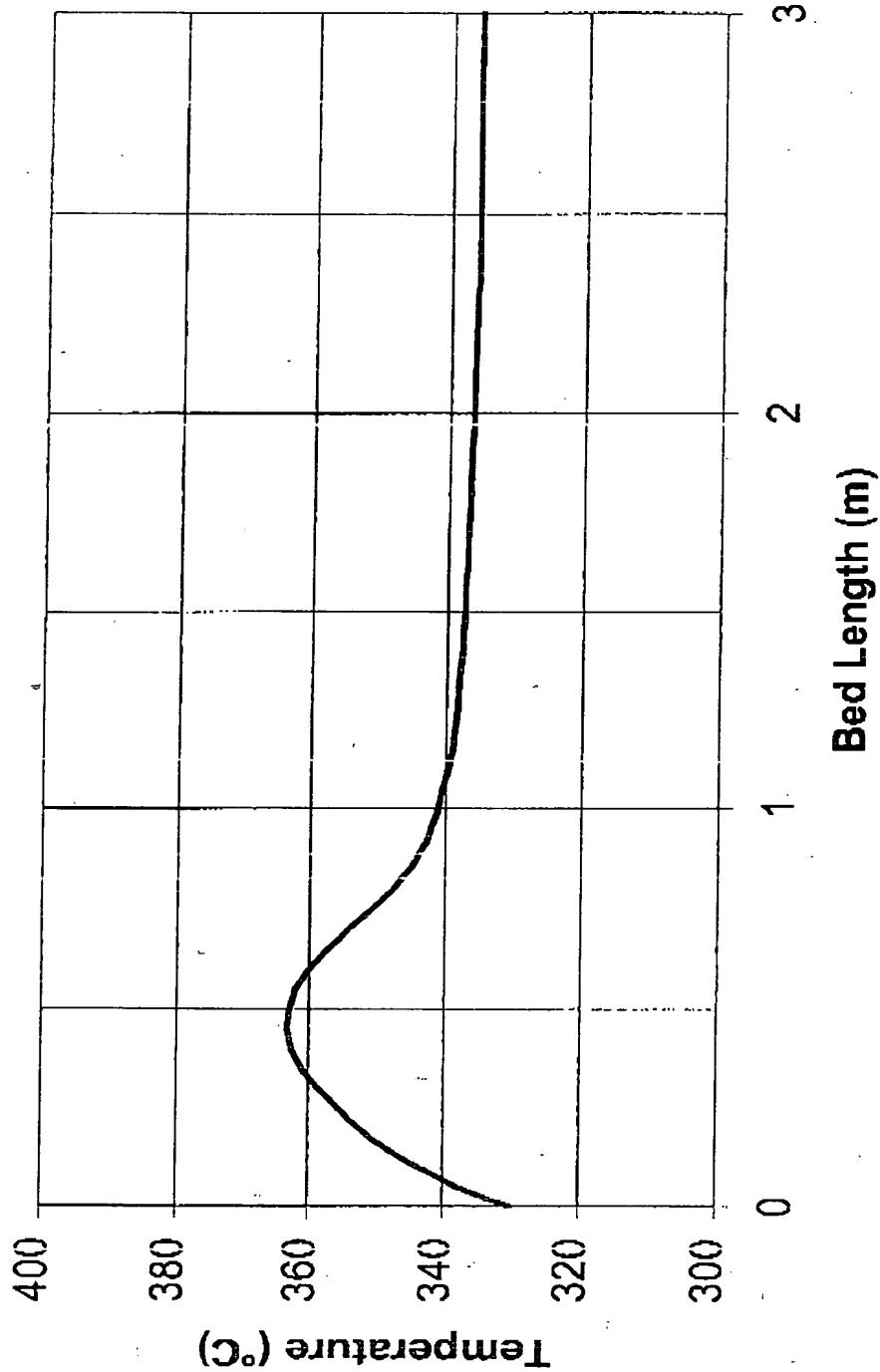
13. JUL. 2000 16:22

NO. 6678 P. 21

2/4

FIGURE 2

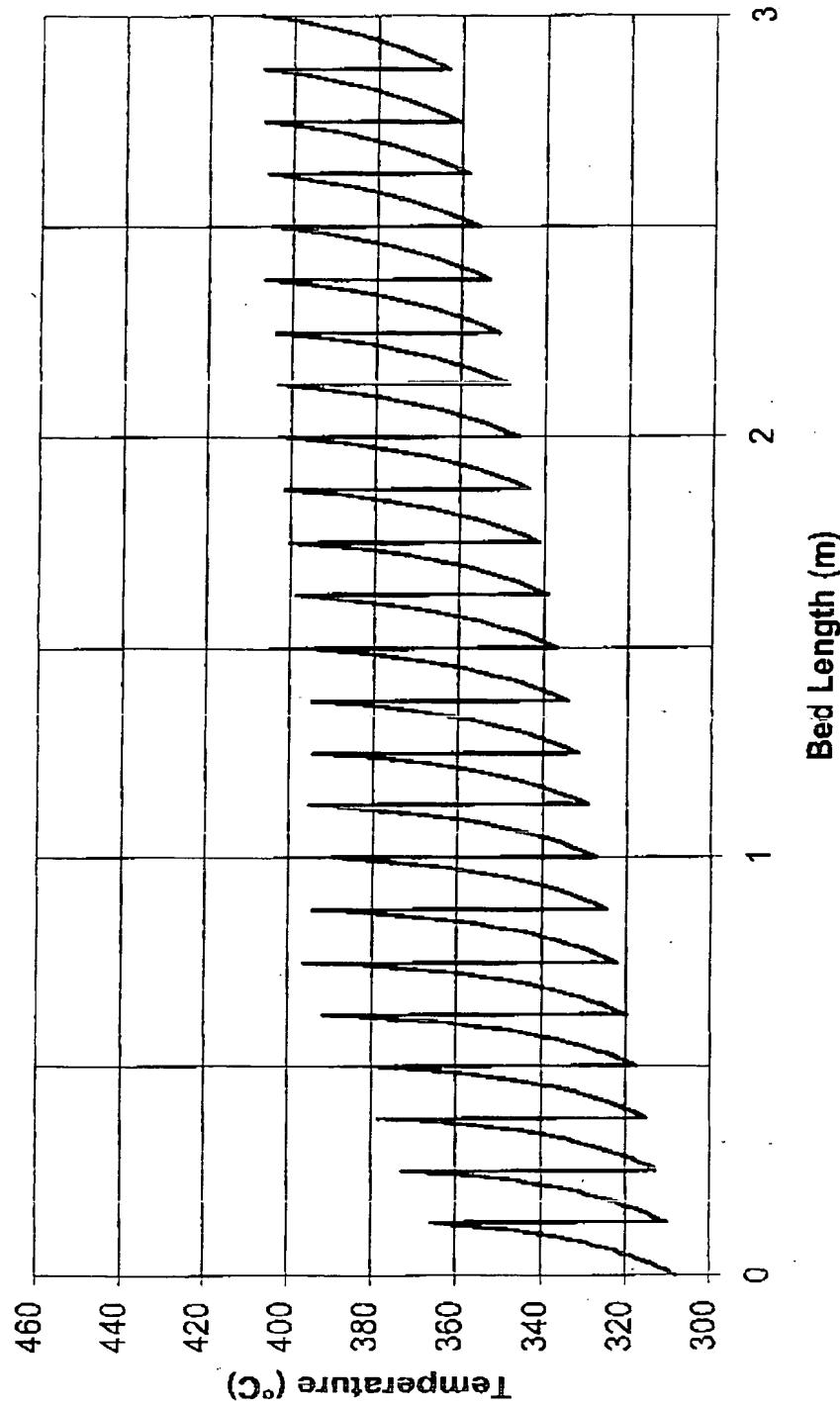
PHTHALIC ANHYDRIDE TUBULAR REACTOR TEMPERATURE PROFILE
(Ortho xylene feed <50 g/Nm³)



3/4

FIGURE 3

PHTHALIC ANHYDRIDE PCR BED TEMPERATURE PROFILE
(Ortho xylene feed >100 g/Nm³)



4/4

FIGURE 4
PHthalic Anhydride PCR

